Recent Multiphase Atmospheric Chemistry Investigations: HCCT-2010 and CAPRAM Developments

Hartmut Herrmann[1], Dominik van Pinxteren[1], Konrad Müller[1], Laurent Poulain[1], Yoshiteru Iinuma[1], Wadinga Fomba[1], Andreas Tilgner[1], Peter Bräuer[1], Taehyoung Lee[2], Jeff Collett[2], Lisa Whalley[3], Dwayne Heard[3], Johannes Schneider[4], Stephan Borrmann[4], Barbara D'Anna[5], Christian George[5]

[1]Leibniz Institute for Tropospheric Research (IfT), Leipzig, Germany
[2]Colorado State University, Department of Atmospheric Science, Fort Collins, USA
[3]University of Leeds, National Centre for Atmospheric Science, Leeds, Great Britain
[4]Max Planck Institute for Chemistry, Particle Chemistry Department, Mainz, Germany
[5]University Lyon; CNRS, UMR5256, IRCELYON, Villeurbanne, France

We have undertaken a complex ground-based chemical aerosol-cloud interaction experiment in autumn of 2010 with international participation. The experiment idea is outlined and first results are described from chemical offline (aerosol impactor and filter samplers, one, three and fives stage cloudwater collectors) as well as from four AMSs deployed in a Lagrangian set-up, consisting of one upwind station, one in-cloud station, and one downwind station in the area of the Schmücke, Germany. During appropriate cloud events, the upwind aerosol composition serves as a reference to study cloud processes at the in-cloud site, as well as possible aerosol modifications at the downwind site. A large number of compounds was analyzed from the offline samples, including inorganic ions, carbon sum parameters, mono- and dicarboxylic acids, sugars and sugar-related compounds, organic carbonyl compounds, metals and metal ions. The AMS measurements allowed for highly time-resolved insights into changes of the aerosol chemical composition during a cloud passage.

In the second part, a number of recent developments for CAPRAM are described: A coupling to MCM is underway, there is a new, more complex CAPRAM halogen module (CAPRAM HM2), and, in collaboration with LISA, Paris, a mechanism generator for organic aqueous phase chemistry (GECKO-A) was developed and is operative now. For the development of GECKO-A, a comprehensive database with kinetic data for more than 600 reactions of organic compounds with hydroxyl and nitrate radicals has been created. For missing kinetic data, suitable estimation methods such as structure-activity relationships have been evaluated and implemented in a protocol for automated mechanism construction. The generator has been used to expand the organic chemistry in CAPRAM up to C4 chemistry in the current study. Together with the MCM, a multiphase mechanism with more than 15000 reactions (over 11000 reactions in the gas phase and over 3600 in the aqueous phase) was used in the current studies. Results from these developments are described. Besides investigations of the concentration profiles of key compounds, detailed time-resolved flux analyses of the source and sink fluxes of those compounds have been performed. The study provided a broader knowledge on the production and destruction of organic compounds, such as dicarboxylic acids, in the tropospheric multiphase system.